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Leaching of technologically enhanced naturally occurring radioactive materials

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Abstract

A form of waste associated with mining activities is related to the type of deposit being mined and to the procedure of exploitation and enrichment adopted. The wastes usually contain relatively large amounts of technologically enhanced naturally occurring radioactive materials (TENORM). The TENORM are often stored on the surface. Consequently, they can be leached as a result of interaction with aqueous solutions of different chemical composition. This further leads to pollution of water and soil in the vicinity of the stored wastes.

The paper presents the results of laboratory investigation aimed at quantifying the leaching process of samples originating from uranium dumps and storage reservoirs associated with brine pumped from coal mines. The leaching process was investigated with respect to selected elements: uranium isotopes, radium isotopes, iron, barium and sodium. The samples were exposed to aqueous solutions of different chemical composition. The experiments revealed that TENORM in form of sulphate compounds are the most resistant against leaching. The leaching coefficient for radium isotopes varies from a few thousandth percent to a few hundredth percent. On the other hand, for TENORM occurring in sand or sludge, the leaching coefficient for uranium and radium isotopes ranged from a few hundredth percent to a few percent.

Keywords: Mining waste; Technologically enhanced naturally occurring radioactive material; Leaching coefficient; Aqueous solutions; Chemical composition

1. Introduction

One of the most important problems in remediation of wastes with occurrence of technologically enhanced naturally occurring radioactive materials (TENORM) is the assessment of vulnerability of radioactive elements contained in these materials to leaching in interactions with aqueous solutions of different chemical composition. This problem was discussed by several authors. For instance, Rajaretmen and Spitz (2000) concluded that leaching coefficient of ^{226}Ra present in complex compounds of sulphates was independent of the chemical composition of interacting aqueous solutions and its average value was assessed to be equal to 1.3%. Chałupnik (2002) investigated leaching of sediments derived from brine of the Upper Silesia coal basin. He showed that leaching coefficients of radium isotopes contained in sludge sediments were significant; they varied between 1% and 10%. Bahranowski et al. (1999) performed leaching of Ca, Mg, K, Na and Fe contained in fly ash deposits resulting from burning of brown and hard coal, which were exposed to water and solution of nitric acid. They found that leaching coefficients for calcium and sodium amounted to 30%, whereas for magnesium and potassium they were significantly smaller, reaching only a few percent. Iron turned out to be the most resistant and it was practically not leached. These authors also concluded that leaching of the investigated elements was most intense when the fly ash interacted with solutions of pH less than 1.

The form in which TENORM usually occur depends on the type of deposit and the procedure of its exploitation. For instance, in case of uranium exploitation TENORM usually occur as marl or shale (Fermandes et al., 1996;

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Jakubick and Hagen, 2000; Piestrzynski et al., 2001). For oil or coal mining, they may have a form of scale or sludge (Rajaretmen and Spitz, 2000; Veguera et al., 2002; Chałupnik et al., 2001). In copper mining they usually occur as shale (Piestrzynski, 1989). Each form of TENORM may behave differently when interacting with aqueous solutions of different chemical properties. Investigations of the leaching coefficient are typically performed as part of remediation projects for sites with technologically enhanced naturally occurring radioactive materials.

The paper presents laboratory investigations of the leaching coefficient for uranium isotopes (^{238}U, ^{234}U), radium isotopes (^{226}Ra, ^{228}Ra) and also some other elements such as barium, iron and sodium which often occur together with TENORM. The investigated samples represent uranium deposit waste named SP-6 originating from the uranium mine open pit at Kletno area, and sludge sediments associated with sedimentary reservoirs (Rontok and Bojszowy) used to clear water and brine pumped from coal mines. The sites are located in the Lower and Upper Silesia Coal Basins, respectively, both situated in the SW region of Poland (see Fig. 1). Detailed description of the sites can be found in Waclawek et al. (2000) and Chałupnik et al. (2001).

2. Investigated materials

Kletno region is the one of the locations in the Lower Silesia Coal Basin, where uranium deposits were exploited in the 1950s. Mining activities were terminated in late 1960s. In this region TENORM occur as marl and shale of grey or orange colour (Piestrzynski et al., 2001).

The Rontok reservoir is a natural pond which is connected with Vistula River by a channel of about 100 m length. Brine pumped from the coal deposit of Silesia colliery are directed to the Rontok reservoir. They are classified as type-A brine. They contain elevated concentrations of barium ions (ca. 1.5 g/L) but no sulphate (Lebecka et al., 1986). The water is pumped from the mine and mixed with surface water, which is usually rich in sulphate ions. This leads to massive precipitation of barium (radium) sulphate compounds appearing as a dense scale. The pumped mine water also carries suspended sediments (fine pieces of coal and clay) which are deposited in the reservoir, making a thick sludge layer.

The Bojszowy reservoir is an artificial pond connected with Vistula River through Gostynka River. Groundwater from the coal deposits of Piast and Czeczot collieries is pumped into this reservoir. The water is classified as type-B brine, rich in sulphate ions (ca. 3.5 g/L) but very poor in barium (Lebecka et al., 1986). Therefore, precipitation of barium and radium ions does not occur when brine is mixed with surface water. Similarly, as in the case of the Rontok reservoir, there is a thick deposit of sludge sediment on the bottom of the Bojszowy pond, made of fine pieces of coal and rock carried out by the pumped water. The average concentrations of ^{226}Ra and ^{228}Ra in water of type A and B are equal to 62.76, 34.67 and 3.45,

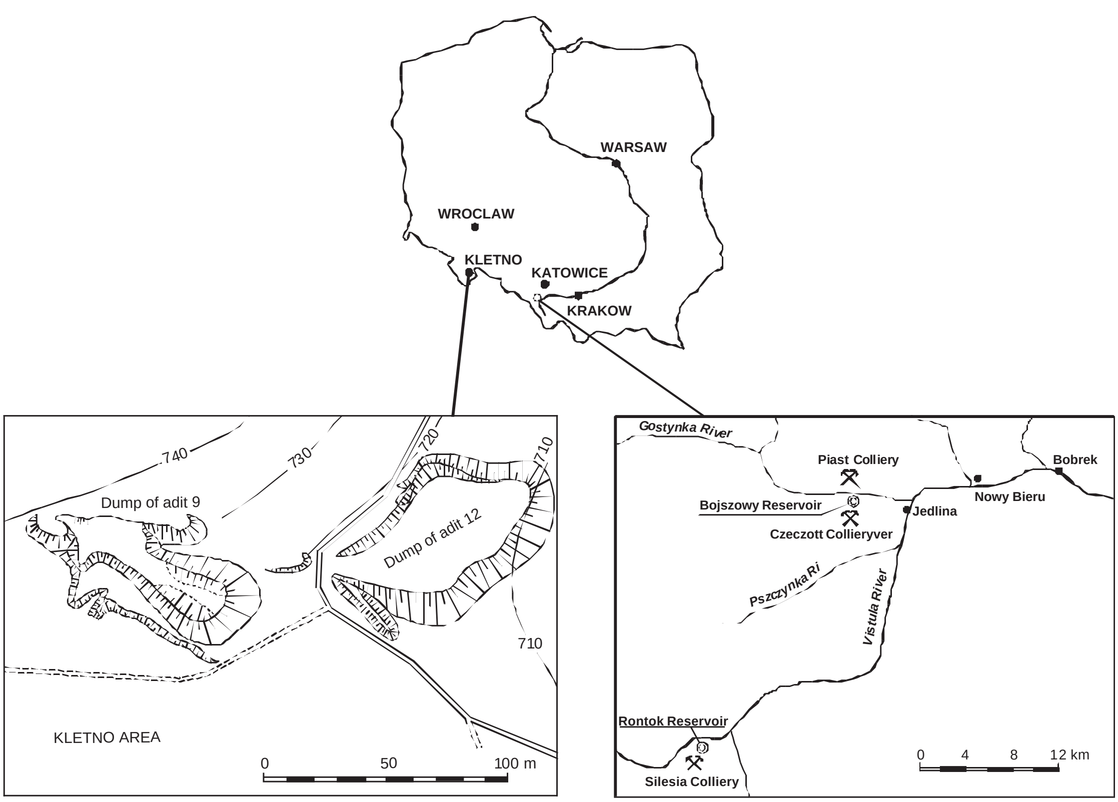

Fig. 1. Sketch of the Kletno area and Upper Silesia Basin.